Low Pressure RO Membrane Desalination of Agricultural Drainage Water

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ABSTRACT

Agricultural drainage water is a complex mixture of dissolved and suspended chemical species and may contain a wide variety of micro-organisms. The application of membrane systems for desalination of agricultural drainage (AD) water requires careful consideration of feed water quality, suitable membrane selection and operating conditions. In order to evaluate the potential applicability of low pressure reverse osmosis (RO) to the treatment of AD water, a diagnostic approach to membrane selection and process evaluation was undertaken in support of a pilot field study in the California San Joaquin Valley. Five candidate membranes were evaluated in a diagnostic laboratory membrane system which provided an initial selection based on salt rejection and product water flux performance for model salt solutions of univalent and divalent cations. Biofouling potential of the selected membranes was also evaluated using two standards strains of bacteria. Preliminary pilot plant performance, based on the selected membranes, was encouraging and has provided the basis for long-term pilot plant testing at higher recoveries to assess the impact of fluctuating AD water feed composition.

KEYWORDS

Agricultural drainage water; reverse osmosis; membrane diagnostics, desalination; biofouling.

1. INTRODUCTION

Tile drainage of irrigated lands is practiced in many semi-arid agricultural regions. Adverse geological conditions in such areas often involve impervious layers underlying fertile land [1,2,3]. Artificial drainage is practiced in order to prevent water-logging and salinity buildup in the root zone of crops. Hydrologic and environmental impacts of artificial drainage have been extensively reviewed by Skaggs *et al.* [4].

The fertile semi-arid California San Joaquin Valley was one of the first regions to install tile drainage systems for irrigation water which has proved most effective approach of controlling root zone salinity. Since the early 1970's, serious consideration has been given to systems for reclamation and reuse of agricultural drainage water. Motivation for application of this technology arose from two major issues. First, a successful reclamation facility would help to augment diminishing supplies of imported irrigation water. Secondly, volume reduction of environmentally hazardous drainage water could also be achieved. The feasibility of reverse osmosis (RO) for drainage water reclamation was first demonstrated in 1971 at the historic pilot facility at Firebaugh California [5,6]. A larger and considerably more sophisticated treatment plant was completed, in the nearby town of Los Baños [7,8,9] in the mid 1980's, to study a variety of operating parameters and to assess the economic feasibility of drainage water reclamation with RO technology as an important component. This plant was unfortunately shut down in 1987 due to concern with high concentrations of selenium in the form of SeO₄²⁻ ion found at Kesterson - the site of a low-lying basin for all tile drainage in that region. Tile drainage in the West Central San Joaquin valley has since been terminated, resulting in a severe hardship for the farming community. If not resumed, a gradual salinity build-up will necessitate the "retirement" of large areas of fertile agricultural land.

A search for solutions to the drainage problem is presently underway, and again, membrane desalination has been given serious consideration owing, in part, to a new generation of high performance low pressure RO and nanofiltration (NF) membranes developed during the last decade [10,11]. Low pressure RO membranes can operate at remarkably low pressures with excellent product water flux and reasonably high levels of salt rejection. However, selection of the appropriate membrane for AD water desalination must involve careful consideration of feed water quality.

Consideration of water quality in relation to optimization of the desalination process is especially critical with AD water, which is a complex mixture of dissolved and suspended organic and inorganic components as well as a wide variety of micro-organisms. The acceptable TDS for irrigation water is about 750 mg/L. However TDS of AD water from the San Joaquin Valley varies between 3,000 and 15,000 mg/L and most samples are close to saturation with respect to gypsum (CaSO₄· 2H₂O). Water reuse necessitates desalination of this water to achieve the desired TD level. Moreover, the control of gypsum scale formation is critical to establishing practical field installations of membrane desalination of AD water. Antiscalants, consisting primarily of polyelectrolytes, have met with some success in inhibiting membrane surface scaling by gypsum. Other important aspects affecting membrane performance are colloidal particles and potential microbiological growth.

Optimization of membrane desalination systems for AD water presents a challenge for system designers and plant operators. Of primary concern is membrane selection for this specific task. An assessment may, in part, be based on controlled laboratory experiments, but overall suitability can be determined only by long-term operation in the field. In addition to membrane selection, the designer must be concerned with operating parameters and appropriate feed water pretreatment systems. Prefiltration can be effective in reducing the problem of

colloidal and biofouling. Design of such pretreatment systems is as important as proper choice of the membrane itself.

In the present work we present laboratory and pilot plant investigations to evaluate the feasibility of membrane desalination of agricultural drainage water in the California San Joaquin Valley. The first objective was concerned with selection of suitable low pressure RO membranes based on testing under carefully controlled laboratory conditions with diagnostic model solutions of the major univalent and divalent cations found in field drainage water. These studies were followed by evaluation of the biofouling potential of the candidate membranes. The second objective was concerned with a critical assessment of membrane suitability and operating conditions, in preparation for a long-term pilot plant operation, based on short-term process data generated from a membrane pilot plant at the Buena Vista Water Storage district in Buttonwillo, California.

2. LABORATORY STUDIES

2.1 Membranes and Materials

Five commercial aromatic polyamide composite low pressure RO membranes were selected from three major manufacturers based on reported ion rejections and flux at a specific applied pressure. These membranes, Hydranautics LFC-1 and ESPA-1 (Oceanside, CA), Dow-FilmTec NF-90 (Minneapolis, MN), and Koch Membrane Systems TFC-ULP and TFC-HR, were stored in accordance with manufacturer specifications. Diagnostic solutions were all prepared using ultra-pure de-ionized water obtained by filtering distilled water through a Milli-Q Water System (Millipore Corp., San Jose, CA). Calcium chloride dihydrate (certified A.C.S), magnesium sulfate (certified A.C.S), sodium chloride (USP/FCC granular), and sodium metabisulfite (certified A.C.S) were obtained from Fisher Scientific (Pittsburgh, PA).

2.2 Membrane Test Unit

A small laboratory plate-and-frame recirculation unit (Figure 1) was used as a diagnostic membrane performance evaluation system. This unit consists of two test cells (Industrial Research Machine Products Co., El Cajon, CA) arranged in parallel with each cell having a flow area of 2.6 cm x 7.6 cm (membrane area of 19.76 cm²) and channel height of 0.266 cm.). The magnetically stirred polyethylene reservoir accommodates up to 18 liters of feed water. A refrigerated recirculator (model 625, Fisher Scientific, Pittsburgh, PA) maintained constant reservoir temperature. A positive displacement pump (Hydra-Cell, Wanner Engineering, Minneapolis, MN) delivers up to 1.1 gpm of feed solution. All membrane performance experiments were conducted at a cross flow velocity of 40 cm/s (corresponding to a Reynolds number of 1,336, based on the channel height). A back-pressure regulator (US Paraplate, Auburn, CA) served to adjust the applied transmembrane pressure. A digital flow meter (model 1000, Fisher Scientific, Pittsburgh, PA), interfaced with a PC, provides for continuous monitoring of permeate flux and accumulated volume. Permeate conductivity, at different times during operation of the unit, is measured using a conductivity meter (model WD-35607-30, Oakton Research, Vernon Hills, IL).

2.3 Membrane Rejection, Flux, and Biofouling Potential

A performance testing protocol for each of the pre-selected membranes was carried out at a fixed temperature of 20°C and applied transmembrane pressure of 100 and 200 psi. Feed solutions consisted of aqueous solutions of 0.05, 0.10, and 0.15 M sodium chloride (NaCl) and 0.01, 0.02, and 0.05 M calcium chloride (CaCl₂), respectively. Concentrations of sodium and calcium chloride were chosen based on selected analytical values of drainage water samples (Table 1 and 2) at the Buena Vista Site in the San Joaquin Valley.

Steady-state conditions for both membrane compactions and the diagnostic experiments were typically achieved within a period of 2-6 hours. The system was operated in a total recycle mode whereby the permeate and concentrate were returned to the feed reservoir. In addition to on-line conductivity and flux measurements, permeate samples were collected at various intervals and returned to the reservoir following the completion of conductivity or ion-specific measurements.

Membrane biofouling potential was evaluated using the biofouling potential assay developed by Ridgway and co-workers [12,13] at the Orange County Water District Biotechnology Laboratory. The assays were performed for each membrane with three reference membranes included to account for potential experimental variations. The three reference membranes were the new and old versions of a fully aromatic cross-linked polyamide FT-30 RO membrane from the Dow Chemical Company (Midland, MI), and a low-pressure cellulose acetate membrane devoid of any post-synthesis surface treatment from Applied Membranes, Inc. (San Marcos, CA). The test bacteria were a hydrophobic strain of Mycobacterium (BT12-100) and a hydrophilic strain of Flavobacterium (PA-6) both radio-labeled with Na₂³⁵SO₄. Two sets of biofouling assays were performed. In the first set, the membranes were contacted in a glass flask (in a shake bath) containing NPM (sodium phosphate + magnesium chloride) buffer and the test bacteria for 5 hours at 28°C. In the second set, the NPM buffer was replaced by actual AD water. The bacterial attachment count (i.e., number of bacteria/cm²) was determined by a LKB Rackbeta 1219 liquid scintillation counter (LSC; Wallac, Gaitherberg, MD).

3. MEMBRANE SELECTION

Membrane selection for AD water desalination was based on initial performance evaluation (salt rejection and permeate flux) at 100 psi transmembrane pressure and subsequent

screening analysis of biofouling potential. Final selection was based on performance testing at 200 psi transmembrane pressure.

Sodium and calcium rejection, R, defined as $R=100~(1-C_p/C_f)$, where C_p and C_f are the solute concentrations in the permeate and feed streams, respectively, by the five selected membranes, at 100 psi trans-membrane pressure and 0.1 N NaCl and $CaCl_2$ solutions, is shown in Figure 2a. As expected, these membranes consistently demonstrate a higher salt rejection and permeate flux for the solutions of the divalent calcium ion than for the univalent sodium ion. This behavior is consistent with published studies for multivalent electrolytes [14,15]. Permeate flux, shown in Figure 2b, ranged from 2 to 9.5 gfd for the NaCl feed solution and 4.8 to 13.6 for the $CaCl_2$ feed solution, with the flux generally decreasing with increased rejection. Membrane NF-90, which exhibited the lowest rejection out of these five membranes, was eliminated from further testing because its performance was below the minimum desired for the expected feed concentrations in the field.

Rejection of calcium and sodium over the range of concentrations expected in the field (over the course of the drainage season) revealed that higher feed concentrations caused a decrease in sodium rejection but had a less pronounced effect on calcium rejection (Figure 3). It is also clearly shown in this figure that, over the range of concentrations of interest and for the individual salt solutions, these low pressure RO membranes provided a higher rejection of calcium compared with sodium. For both NaCl and CaCl₂ feed solutions, the permeate flux decreased with increasing feed concentration for the respective salt solutions (Figure 4). The TFC-ULP membrane, which showed the lowest salt rejection, exhibited the highest permeate flux for all feed compositions tested. In general, experimental results confirmed the fact that membrane selection involves a trade-off between solute rejection and permeate flux. We also note that in the present diagnostic evaluation, the "true" recovery was not directly evaluated due

to the small membrane surface areas in the laboratory RO system. However, the measured permeate flux and an overall salt rejection can be used to estimate recovery for full-scale operation.

While the first set of experiments at 100 psi transmembrane pressure provided an initial baseline for membrane performance evaluation, the next step in screening the remaining candidate membranes was accomplished by membrane biofouling potential analysis. This analysis was conducted at the Orange County Water District laboratories. Analysis of filtered AD water samples revealed a bacterial count of 1.36 x 10⁶ bacteria per mL. This measured bacterial count is at a level of concern over a long term of operation and thus suggested the evaluation of membrane biofouling potential. This analysis, carried out in buffer solutions (Figure 5), revealed that the LFC-1, TFC-ULP and TFC-HR membranes displayed the lowest biofouling potential, while the ESPA-1 and NF-90 membranes had the highest biofouling potential. In order to confirm these results, the same biofouling assay was repeated with selected membranes using the filtered AD water. The results, as shown in Figure 6 clearly indicate the trend observed with buffer solutions containing test bacteria.

In the subsequent membrane selection step, performance of the lowest biofouling membranes (LFC-1, TFC-ULP and TFC-HR) was evaluated at a higher transmembrane pressure of 200 psi in order to approximate the higher range of operating pressure in the pilot plant study. As expected a higher rejection (Figure 7) and flux (Figure 8) were obtained relative to the data collected at 100 psi. Divalent calcium ions were rejected to a greater extent than sodium ions in both pressure ranges. All three membranes exhibited rejections above 90% for all three feed concentrations of calcium and sodium chloride except for 89% sodium rejection for the TFC-ULP membrane using 0.15 N NaCl feed. It should also be noted that the LFC-1 and TFC-HR

membranes demonstrated greater than 99% rejection for all feed concentrations of calcium chloride. The above three membranes were all suitable candidates for the pilot plant field study.

4. PILOT PLANT STUDY

The pilot plant, designed and operated by Boyle Engineering, was located at the Buena Vista Water Storage District in Buttonwillow, California. The plant, shown schematically in Figure 9, consisted of a pretreatment multi-media filtration system and a two-stage portable reverse osmosis unit. Pre-filtration consisted of three garnet filters and a sand filter. During operation, alum and scale inhibitor Hypersperse AS20 obtained from GE Betz (Trevose, PA) were both injected into the feed water at a rate of 5 mg/l. Acid was also injected to adjust the feed water pH to 6.8 in order to further reduce potential calcium carbonate scaling. The plant was designed to handle a water feed flow rate of up to 27 gpm. The system was configured with six pressure vessels each containing three spiral-wound membrane elements, arranged in a 2:2:1:1 array with a total membrane surface area of 133.2 m² (1440 ft²).

Membranes considered for the pilot study included the TFC-ULP and TFC-HR. The TFC-ULP membrane displayed higher flux relative to the other low-fouling candidate membranes (Figures 4 and 8). This membrane had an acceptable rejection for calcium over the concentration ranges of interest and performed reasonably well for sodium at the mid- to low concentration range (Figure 3). The overall rejection performance of this membrane was lower than that measured for the other candidate membranes. The TFC-HR membrane had the best rejection performance (>93%) for all conditions tested but had a significantly lower flux compared to the TFC-ULP membrane. Since calcium removal is of paramount importance for AD water, the LFC membrane was not selected since measured flux, for the calcium solutions (Figs. 4b and 8b) was lower relative to the TFC-HR and TFC-ULP membranes.

In order to evaluate anticipated performance in the pilot plant facility a process screening analysis was carried out, using the POPRO6 software (Koch Membrane Systems), for different configurations involving TFC-HR and TFC-ULP membrane modules (80 ft² per module). The analyzed configurations included: (a) stage 1: TFC-ULP, stage 2: TFC-ULP; (b) stage 1: TFC-ULP, stage 2: TFC-HR; (c) stage 1: TFC-HR, stage 2: TFC-ULP; and (d) stage 1: TFC-HR, stage 2: TFC-HR. Input variables included temperature, pH, feed water composition, feed flow rate, percent recovery, number of pressure vessels, number of membrane elements per pressure vessel, type of membrane, and fouling allowance for the membranes. Assuming preventive measures are taken to reduce fouling and scaling, the fouling allowance (expressed as percent loss of net transmembrane pressure) was set at 15 percent for this analysis. Feed water composition was set as in Tables 1 and 2, respectively, and feed water pH and temperature were set to 6.8 and 22°C, respectively. The desired operation was set at a feed flow rate of 20 gpm with a target product recovery of 50 percent, with an overall permeate flow rate of 10 gpm [16]. Several assumptions were made in these screening simulations: the permeate backpressure and interbank pressure loss were neglected and the impact of antiscalants and acid (to minimize calcium carbonate scaling) were not directly considered.

The simulation results revealed that for the specified recovery and permeate flow, the percent TDS rejection for the four configurations a-d were 96.3, 96.8, 97.3 and 97.8, respectively, with the corresponding inlet pressures of 127.2, 131.1, 151.4 and 165.4 psi. The ratio of stage 1 to stage 2 permeate flux was approximately 1.1 for configuration c while it ranged from 1.8-5.75 for the other three configurations tested. Although all configurations yielded a high rejection level, having both stages operate at a similar level of permeate flux, as obtained in configuration c, was sought as the preferred operating condition while maintaining a reasonably low transmembrane pressure. For all configurations, the process analysis results

revealed that the concentrate was at the saturation level with respect to calcium sulfate and about 24 times the saturation level for barium sulfate. These oversaturation levels are indeed of concern and thus suggest the need for additional pretreatment considerations.

The pilot plant was operated from period of August 1, 2000 to September 13, 2000. During plant operation, feed TDS varied from 3,500-8,800 mg/L, similar to the concentration range covered in the diagnostic laboratory-scale membrane screening study. The plant was operated at a feed pressure of 145-235 psi. In the initial operation of the facility ESPA-1 membrane was used for both stages since it was available and already installed from a previous pilot plant study. However, after the first 530 hours of operation, the TFC-HR membrane was installed in the first stage and after operating for 720 hours the TFC-ULP membrane was installed in the second stage

During the first 530 hours feed conductivity varied from 5080 to 8770 microsiemens (mS)/cm and percent rejection remained in the range of about 84-91 % (Figure 10). The percent recovery and normalized flux fluctuated by up to 78% for percent recovery and 220% for normalized flux (Figure 11). It should be noted that a rejection level of 91% would be at the limit of treating a feed TDS of 8333 mg/L while meeting the requirement of a 750 mg/mL TDS product for agricultural water application. Clearly, higher TDS levels often encountered in AD water would require a higher rejection. After installation of the TFC-HR membrane in the first stage ((New 1st stage membrane on Figure 11) rejection increased and was sustained at a level above 90%. Although some decline in recovery and normalized flux was observed, these were re-established once the ESPA-1 membrane in the second stage was replaced (New 2nd stage membrane on Figure 11) by the higher flux lower biofouling TFC-ULP membrane. During the period following the replacement of stage 2, feed conductivities decreased significantly (down to ~4300 ms/cm), which along with the performance characteristics of TFC-HR, allowed salt

rejection to reach 95%. Following the replacement of stage 2 membranes, the RO system was also able to achieve relatively stable normalized flux (an average of 0.09 GFD/psi) and recovery of about 50 %.

While the above field study results are preliminary and conducted over a period that was limited by the unusually short drainage season, the overall performance is encouraging and suggests further evaluation of membrane desalination for agricultural drainage water. Current efforts are focused on a longer field study designed to evaluate longer-term performance at higher product water recovery.

6. CONCLUSIONS

The present study presents an approach to evaluating membrane desalination of agricultural drainage water based on the combination of a laboratory diagnostic study with a pilot field evaluation. Two different high performance RO membranes were installed in the two stage plant with a designed product water output of approximately 50,000 gallons per day. The plant provided product water recovery on the order of 50% from a feed water salinity range of ~ 3500-8800 mg/L TDS. The first stage pilot plant performance evaluation was relatively short due to a diminished flow of drainage water during the 2000 year irrigation season. Despite the short duration of this preliminary field study, the feasibility of effective agricultural drainage water desalination with high performance low pressure RO membranes was clearly demonstrated in this cooperative effort between government, industry and academia.

The study points out that the selection of suitable membranes for desalination of agricultural drainage water requires membrane characterization with regard to flux, ion rejection, biofouling potential and propensity for scale formation. The choice of strategies for reduction of fouling due to both mineral scale and micro-organisms can, in principle, involve suitable

filtration pretreatment, use of chemical additives and appropriate membrane selection. In the present study, it was shown that when selecting a membrane, one may have to consider the trade-offs between reduction in biofouling potential and membrane performance as well as between membrane salt rejection and permeate flux. Work presently underway involves expanded field testing and the evaluation of strategies for reducing membrane fouling via the combination of pretreatment strategies and optimization of membrane system configuration, all specifically targeted for desalination of agricultural drainage water in the California San Joaquin Valley.

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LIST OF TABLES

- **Table 1.** Basic properties of typical Buena Vista drainage water
- **Table 2**. Concentrations of major ions in Buena Vista drainage water

LIST OF FIGURES

- Figure 1. Laboratory-scale RO/NF membrane system.
- **Figure 2.** Comparison of rejection (a) and permeate flux (b) for five commercial low pressure RO membranes (transmembrane pressure= 100 psi, $T = 20^{\circ}\text{C}$).
- **Figure 3.** Comparison of percent rejection for four candidate membranes, using (a) different NaCl feed concentrations; and (b) different CaCl₂ feed concentrations. (Transmembrane pressure= 100 Psi, Temperature= 20°C).
- **Figure 4.** Permeate flux comparison for four candidate membranes using (a) different NaCl feed concentrations; and (b) different $CaCl_2$ feed concentrations. (Transmembrane pressure= 100 psi, Temperature= $20^{\circ}C$).
- **Figure 5.** Comparison of biofouling potential for selected membranes. CA, FT-30 (n), and FT-30 (o) are membranes used as controls. **NPM solution** (10 mM sodium phosphate + 1mM MgCl₂, pH 7.0) was used as buffer. A hydrophobic strain of Mycobacterium and a hydrophilic strain of Flavobacterium were used as the test bacteria. Bacterial attachment (B/F) is the ratio of bacterial count on the membrane to the number of free bacteria in solution.
- **Figure 6.** Comparison of biofouling potential for candidate membranes. CA, FT-30 (n), and FT-30 (o) are membranes used as controls. Note: **Buena Vista water** was used without buffer. A hydrophobic strain of Mycobacterium and a hydrophilic strain of Flavobacterium were used as the test bacteria.
- **Figure 7.** Comparison of percent rejection for four candidate membranes, using (a) different NaCl feed concentrations; and (b) different $CaCl_2$ feed concentrations. (Transmembrane pressure= 200 Psi, Temperature= $20^{\circ}C$).
- **Figure 8.** Permeate flux comparison for four candidate membranes using (a) different NaCl feed concentrations; and (b) different $CaCl_2$ feed concentrations. (Transmembrane pressure= 200 psi, Temperature= $20^{\circ}C$).
- Figure 9. Process flow diagram of pilot plant
- **Figure 10.** Measured feed, concentrate, and permeate conductivities of the RO system during operation of the pilot plant.
- **Figure 11.** Percent rejection based on measured conductivites, percent recovery, and normalized flux of the RO system during the operation of the pilot plant.

 Table 1. Basic properties of typical Buena Vista drainage water

Total dissolved solids (TDS), mg/L	5250
Total organic carbon (TOC), mg/L	2.77
Hardness, mg/L	1630
Turbidity, NTU	0.8

Table 2. Concentrations of major ions in Buena Vista drainage Water

Substance	Concentration	
Cations	(mg/L)	(mol/L)
Na ⁺	1150	0.0500
Ca ⁺	555	0.0139
Mg ⁺	60.7	0.0025
Anions		
Cſ	2010	0.0567
SO ₄ ⁻²	1020	0.0106
HCO ₃	291	0.0048

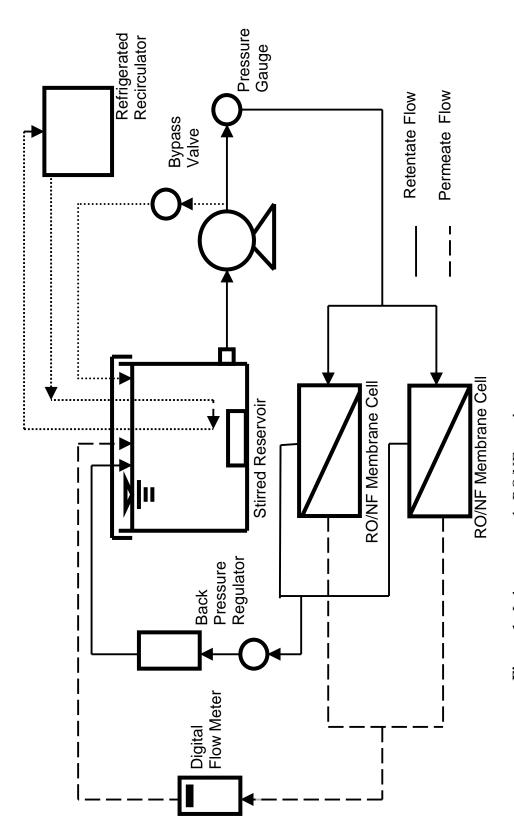


Figure 1. Laboratory-scale RO/NF membrane system.

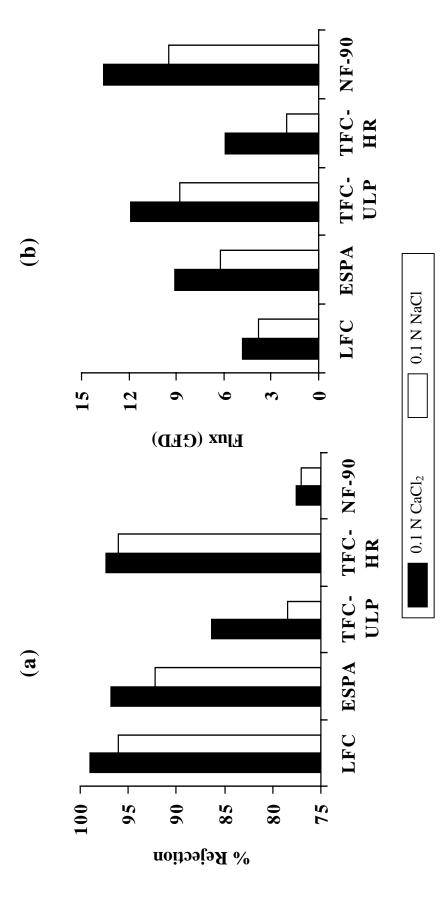
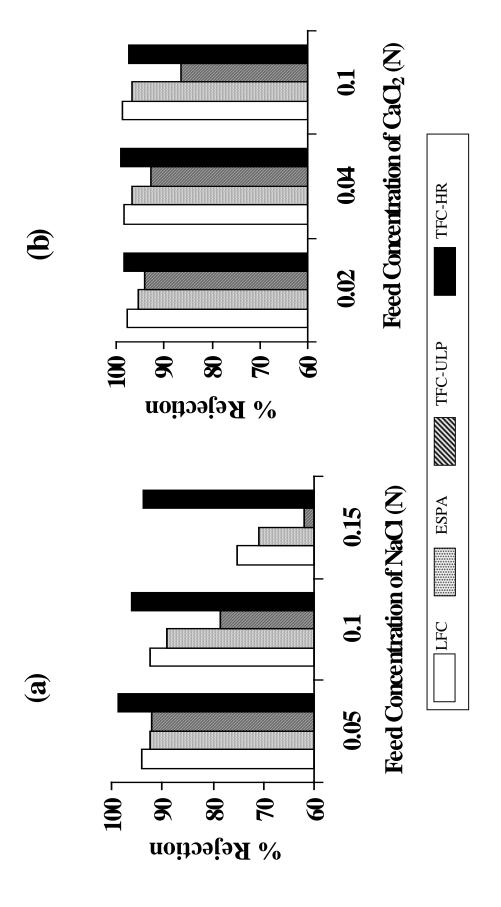


Figure 2. Comparison of rejection (a) and permeate flux (b) for five commercial RO membranes (transmembrane pressure= 100 psi, $T=20^{\circ}\text{C}$).



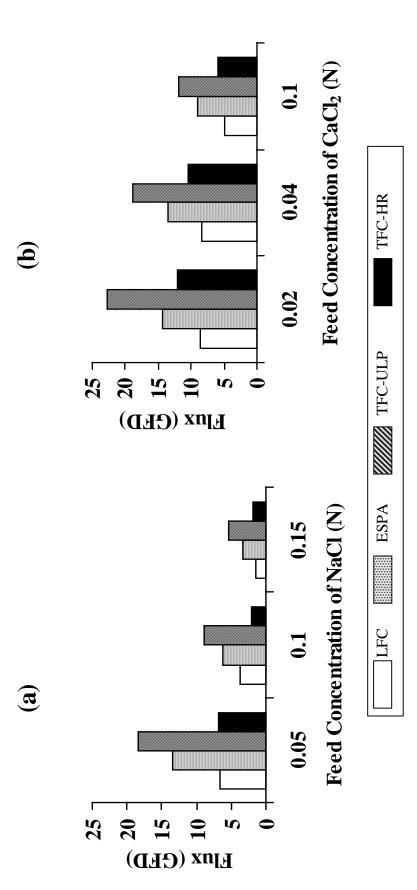
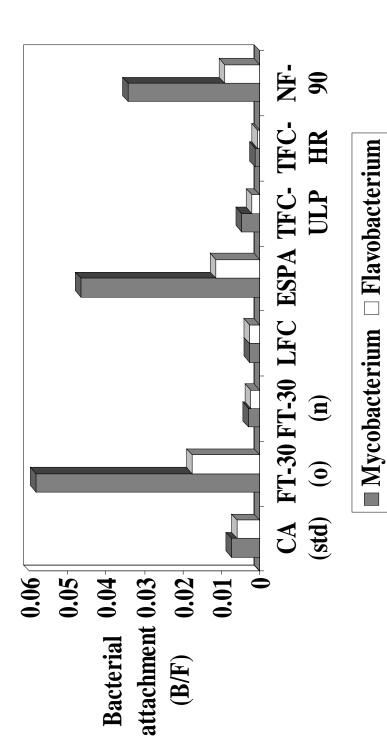
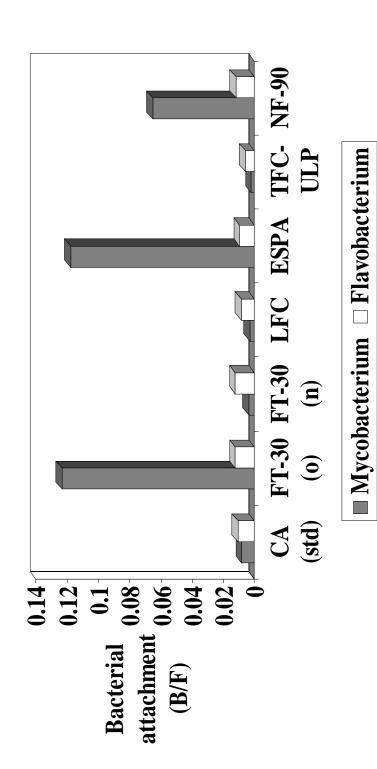


Figure 4. Permeate flux comparison for four candidate membranes using (a) different NaCl feed concentrations; and (b) different CaCl₂ feed concentrations. (Transmembrane pressure= 100 psi, Temperature= 20°C).



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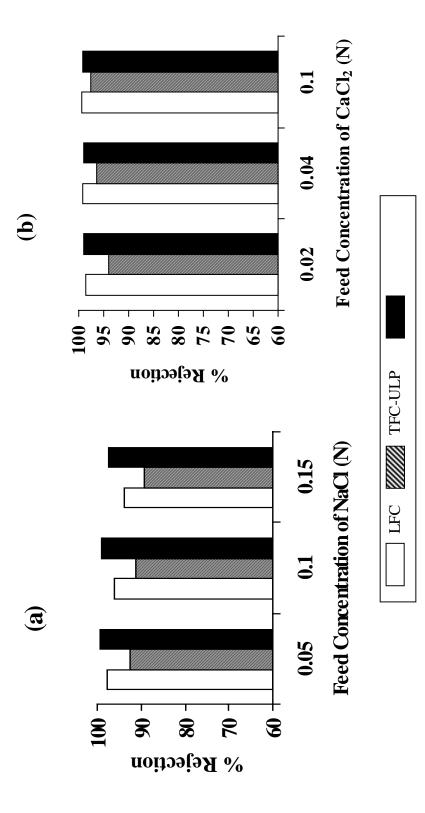


Figure 7. Comparison of percent rejection for four candidate membranes, using (a) different NaCl feed concentrations; and (b) different CaCl₂ feed concentrations. (Transmembrane pressure= $200 \, \text{Psi}$, Temperature= 20°C).

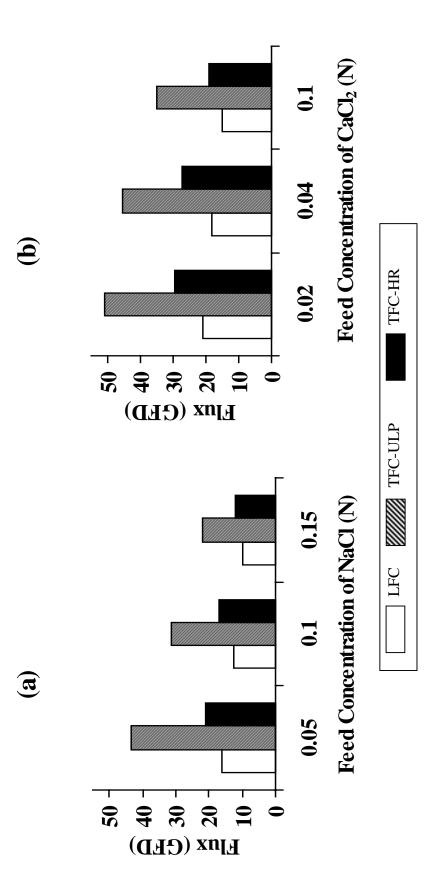


Figure 8. Permeate flux comparison for four candidate membranes using (a) different NaCl feed concentrations; and (b) different $CaCl_2$ feed concentrations. (Transmembrane pressure= 200 psi, Temperature= $20^{\circ}C$).

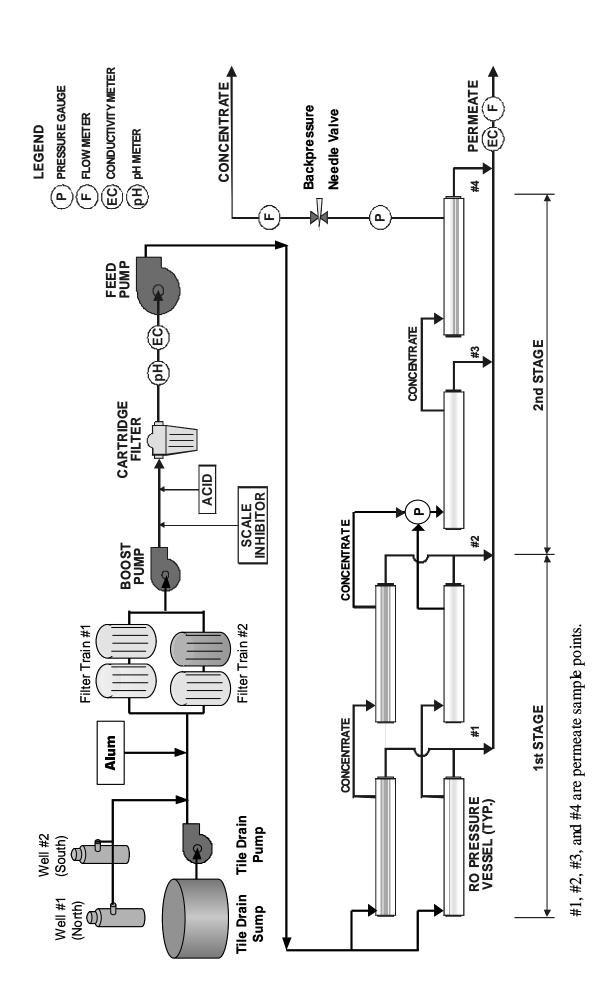


Figure 9. Process flow diagram of pilot plant

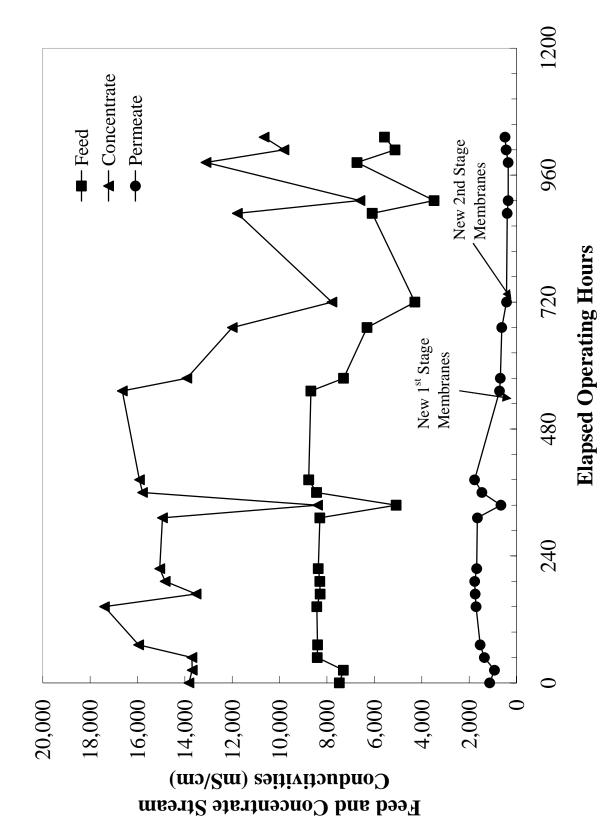


Figure 10. Measured feed, concentrate, and permeate conductivities of the RO system during operation of the pilot plant.

